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## Measurements of an anomalous global methane increase during 1998

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**Abstract.** Measurements of atmospheric methane from a globally distributed network of air sampling sites indicate that the globally averaged CH<sub>4</sub> growth rate increased from an average of 3.9 ppb yr<sup>-1</sup> during 1995-1997 to 12.7 ± 0.6 ppb in 1998. The global growth rate then decreased to 2.6 ± 0.6 ppb during 1999, indicating that the large increase in 1998 was an anomaly and not a return to the larger growth rates observed during the late-1970s and early-1980s. The increased growth rate represents an anomalous increase in the imbalance between CH<sub>4</sub> sources and sinks equal to ~24 Tg CH<sub>4</sub> during 1998. Wetlands and boreal biomass burning are sources that may have contributed to the anomaly. During 1998, the globally averaged temperature anomaly was +0.67°C, the largest temperature anomaly in the modern record. A regression model based on temperature and precipitation anomalies was used to calculate emission anomalies of 11.6 Tg CH<sub>4</sub> from wetlands north of 30°N and 13 Tg CH<sub>4</sub> for tropical wetlands during 1998 compared to average emissions calculated for 1982-1993. In 1999, calculated wetland emission anomalies were negative for high northern latitudes and the tropics, contributing to the low growth rate observed in 1999. Also 1998 was a severe fire year in boreal regions where ~1.3 × 10<sup>5</sup> km<sup>2</sup> of forest and peat land burned releasing an estimated 5.7 Tg CH<sub>4</sub>.

### 1. Introduction

Atmospheric methane (CH<sub>4</sub>) is of considerable interest due to its importance as a greenhouse gas, the role it plays in tropospheric chemistry, and its role in terminating catalytic O<sub>3</sub> destruction cycles involving halogens in the stratosphere. Recent observations of relatively large interannual variations in CH<sub>4</sub> growth rate, superimposed upon a trend that has decreased monotonically for almost 20 years, may provide information to improve our understanding of methane's budget of sources and sinks.

Most anomalies in CH<sub>4</sub> growth rate have been related to large-scale natural events such as the eruption of Mt. Pinatubo during 1991. When assessing interannual variations in CH<sub>4</sub> growth

rate, it is often difficult to determine with certainty which of many possible sources or sinks varied to cause the CH<sub>4</sub> growth rate anomaly. For example, Dlugokencky et al. [1996] showed that SO<sub>2</sub> emitted during the eruption of Pinatubo (and the subsequent sulfate aerosol) affected tropical photochemistry and caused larger CH<sub>4</sub> growth rates in the tropics during 1991. Hogan and Harriss [1994] attributed the decrease in CH<sub>4</sub> growth rate observed at high northern latitudes during 1992 to decreased emissions from northern wetlands due to low temperatures observed as a result of the eruption [Dutton and Christy, 1992], while Bekki et al. [1994] attributed this decrease to increased photochemical destruction of CH<sub>4</sub>. It was also suggested that decreased methane emissions from the fossil fuel sector of the former Soviet Union, brought about by the collapse of the Soviet Union, contributed to the decreased CH<sub>4</sub> growth rate in 1992 [Law and Nisbet, 1996; Dlugokencky et al., 1994a].

Here we present globally and zonally averaged CH<sub>4</sub> mole fractions, and examine the large increase in CH<sub>4</sub> growth rate that was observed during 1998. We suggest that this increased growth rate was due, at least in part, to anomalously high temperatures and precipitation during 1998, which led to greater emissions from high northern latitude and tropical wetland regions. A second, smaller potential contributor was CH<sub>4</sub> emitted from boreal biomass burning, particularly from Siberia during late-summer, 1998.

## 2. Experimental Methods

Our main objective is to make high-precision measurements of the global distribution of atmospheric methane that can be used as a top-down constraint on the global CH<sub>4</sub> budget. The sampling and measurement details have been described elsewhere [Dlugokencky et al., 1994b], so only a brief summary is given here. Samples are collected in duplicate, approximately weekly, from a globally distributed network of background air sampling sites (Figure 1) and analyzed in Boulder, CO, USA for CH<sub>4</sub>, CO<sub>2</sub>, CO, H<sub>2</sub>, N<sub>2</sub>O, SF<sub>6</sub>, and <sup>13</sup>C and <sup>18</sup>O in CO<sub>2</sub>. Methane is determined by gas chromatography with flame ionization detection. The relative precision of the measurements during the period 1991 to 1999 was ~0.1%. All measurements are reported in dry-air mole fraction (nmol mol<sup>-1</sup>, abbreviated “ppb”) relative to an internally-consistent standard gas scale. This scale is propagated from one cylinder to the next with an accuracy of 0±0.2 ppb (95% confidence limit). We are confident that the changes in CH<sub>4</sub> growth rate described here are not the result of measurement artifacts due to drifts or shifts in our standard scale [Steele et al., 1992; Dlugokencky et al., 1994b] or artifacts of our sample collection and storage methods. The measurements are edited for sampling and analytical problems and then selected for background conditions. These quality-control steps assure that the measurements are representative of large well-mixed volumes of the

tropospheric boundary layer. We include a total of 43 time series, excluding from this analysis mountain sites, those sites that are frequently affected by local-scale pollution, and sites with large gaps in their measurement records. Data are available from the CMDL World Wide Web page (path: <ftp://ftp.cmdl.noaa.gov/ccg/ch4/flask/>).

Measurements from each site are fitted by the function

$$f(t) = a_1 + a_2t + a_3t^2 + \sum_{i=1}^4 [a_{2i+2} \sin(2\pi it) + a_{2i+3} \cos(2\pi it)] \quad (1)$$

which approximates the long-term trend (quadratic polynomial) and seasonal cycle (harmonics). The residuals from (1) are calculated and converted to frequency domain, then multiplied by two low-pass filters [Thoning et al. 1989]. One filter captures variability with periods of 45 days and longer, and this result is added to  $f(t)$  to give a smooth curve  $S(t)$ . The second filter captures variability with periods longer than one year, and this result is added to the polynomial component of  $f(t)$  to give the deseasonalized trend,  $T(t)$ . Methane values are extracted at synchronized intervals ( $48 \text{ yr}^{-1}$ ) from smooth curves ( $S(t)$ ) fitted to the data from each sampling site, and another curve is fitted to these values as a function of latitude at each time step [Dlugokencky et al., 1994b]. These latitude slices define an evenly spaced matrix of  $\text{CH}_4$  mole fractions in the boundary layer as a function of time and latitude. The matrix is averaged to define global, hemispheric, semihemispheric (H<sub>NH</sub>=30-90°N; L<sub>NH</sub>=0-30°N; L<sub>SH</sub>=0-30°S; and H<sub>SH</sub>=30-90°S), and polar (P<sub>NH</sub>=53-90°N) zonally averaged values.

Methane emission anomalies from boreal wetlands during 1998 were calculated using an adaptation of the global process-based model of Walter [1998]. Model  $\text{CH}_4$  emissions and soil concentration profiles were tested against 6 comprehensive data sets from different wetlands. Observation periods ranged from 1 season to 3 years [Walter and Heimann, 2000; Walter, 1998]. The adaptation used to calculate anomalies is based on a 12-year run (1982-1993) of the global process-based model. The regression model uses NCEP soil temperature (0-10 cm depth) and precipitation anomalies calculated relative to the 1980-1999 climatology. Wetland distribution and environmental characteristics were from Matthews and Fung [1987] (see Figure 1 for wetland distribution).

### 3. 3 Results and Discussion

#### 3. 1. Methane Observations

Globally averaged  $\text{CH}_4$  mole fractions, deseasonalized trend line ( $T(t)$ ), and instantaneous

growth rate (time-derivative of  $T(t)$ ) are plotted in Figure 2. The anomalous increase in  $\text{CH}_4$  global growth rate during 1998 is one of a series of anomalies superimposed on a long-term decrease in growth rate. The observed long-term decrease in growth rate is consistent with constant  $\text{CH}_4$  emissions and lifetime for a system approaching chemical steady state [Francey et al., 1999; Etheridge et al., 1998; Dlugokencky et al., 1998], or with increasing emissions that, in part, balance a decreasing  $\text{CH}_4$  lifetime [Krol et al., 1998; Karlsdottir and Isaksen, 2000]. Annual increases in  $\text{CH}_4$  (in ppb) by latitude zone were determined from  $T(t)$  as the difference in  $\text{CH}_4$  mole fraction from the beginning to the end of each year (1984-1999), and these are summarized in Table 1. The increase in globally averaged  $\text{CH}_4$  during 1998 was 12.70.6 ppb (uncertainty is 1\_), which represents an imbalance between sources and sinks of 35 Tg  $\text{CH}_4$  (based on the conversion factor in Fung et al. [1991], and where 1 Tg =  $10^{12}$  g). The average rate of increase in methane during the 3 years prior to 1998 was 3.9 ppb  $\text{yr}^{-1}$ , or an increase of 11 Tg  $\text{CH}_4 \text{ yr}^{-1}$  in the global  $\text{CH}_4$  burden; this corresponds to an anomaly in the difference between emissions and sinks of 24 Tg  $\text{CH}_4$  for 1998. An alternative method to calculate anomalous emission is based on the assumption that atmospheric  $\text{CH}_4$  is approaching steady state [Dlugokencky et al., 1998]. Annual  $\text{CH}_4$  increases based on this assumption are listed in the "predict" column in Table 1. Using this approach, the anomaly in 1998 would be 23 Tg  $\text{CH}_4$ .

For the semihemispheres (each representing 25% of the global atmosphere), the largest growth rate during 1998 was observed in the high northern latitudes (30-90°N) at 15.2 1.2ppb. At finer spatial resolution, the 1998 increase for the northern-most 10% of the atmosphere (53-90°N), was 17.20.8 ppb. In the southern hemisphere, the  $\text{CH}_4$  increase at low latitudes (0-30°S) was 14.0 0.8ppb. During 1999, the global increase was 2.6 0.6ppb, comparable to values observed in the three years prior to 1998 (1999 values are preliminary and will change slightly after measurements from 2000 are included in the zonal averages). A slight decrease in  $\text{CH}_4$  mole fraction was observed in the northern hemisphere during 1999, -1.61.1 ppb. This low growth rate is in part due to mixing of "excess"  $\text{CH}_4$  to the southern hemisphere and to higher altitudes, but it also suggests that 1998 was an anomalous year and does not represent a return to larger growth rates observed in the late-1970s and early-1980s.

### 3. 2. Possible Reasons for Increased Growth Rate

In our attempt to understand the growth rate anomaly of 1998, we consider other known anomalies during that year that may have affected  $\text{CH}_4$  emissions or sinks. During 1998, the globally averaged surface temperature anomaly was 0.67°C, for a base period of 1951-1980, the warmest year on record [Hansen et al., 1999; NASA GISS WWW page, path: <http://>

[www.giss.nasa.gov/data/update/gistemp/](http://www.giss.nasa.gov/data/update/gistemp/)]. This suggests that a change in emissions from a source with temperature-dependent emissions possibly contributed. From the CH<sub>4</sub> measurements we know the largest anomaly in growth rate was observed for latitudes north of 53°N. Wetland emissions are sensitive to temperature (~20% increase in emissions for a 1°C increase in the model) and about 20% of natural wetland emissions of CH<sub>4</sub> are from latitudes north of 53°N. Estimates of CH<sub>4</sub> emission anomalies calculated with the regression model are summarized by semihemisphere for 1998 and 1999 in Table 2. The emission anomaly for northern wetlands (>30°N) during 1998 was +11.6 Tg CH<sub>4</sub> compared to the mean emissions calculated for the period 1982-1993. Qualitatively, this fits nicely with the relatively large growth rate observed at 53-90°N. The calculated tropical anomaly was +13 Tg CH<sub>4</sub> with ~90% being emitted in the southern tropics. Again, we have good qualitative agreement with the measurements: for the semihemispheres, the second largest growth rate was observed in the southern tropics (14.0 8ppb). The total anomaly calculated for wetland emissions in 1998 was 24.6 Tg CH<sub>4</sub>, which appears to be in good agreement with the observed anomaly. The good agreement must be viewed with caution. Calculated wetland emission anomalies are upper limits, and they could be overestimated by up to 40%. This is because the estimate of global wetland emissions by the model is ~40% larger than current estimates (e.g., Fung et al. [1991]). The model does not, however, allow for expansion of wetlands due to increased precipitation or melting of permafrost in warm, wet years such as 1998. The important point is that both the observations and the model suggest strong positive emission anomalies during 1998, and the spatial pattern of the anomalies predicted by the model, which we have high confidence in, is in good agreement with the observed spatial pattern of increased growth rate.

Methane is emitted from rice agriculture through the same mechanisms involved in natural wetland emissions. For rice fields, the factors controlling emission rates are altered by management practices such as flooding. Therefore emissions from flooded fields are not affected by precipitation anomalies, unless there is insufficient precipitation to accomplish the flooding. Positive temperature anomalies in the tropics, as observed during 1998, likely would result in increased emissions from rice agriculture [Khalil et al., 1998], but this contribution was not quantified by the regression model.

A second possible anomalous source of CH<sub>4</sub> was from biomass burning in boreal regions. Kasischke et al. [1999] have found that, from 1970-1999, about 80% were light fire years where ~0.7 Tg CH<sub>4</sub> were released from boreal regions. The remaining years, including 1998, were severe fire years. During 1998, ~8.5 x 10<sup>4</sup> km<sup>2</sup> burned in Russia; ~6 x 10<sup>4</sup> km<sup>2</sup> burned after August 1 in the Russian Far East, and the remaining 2.5 x 10<sup>4</sup> km<sup>2</sup> was distributed

throughout the summer fire season (May 1 to 15 October). In Canada,  $\sim 4.5 \times 10^4$  km<sup>2</sup> burned, for a total of  $\sim 13 \times 10^4$  km<sup>2</sup> that burned in boreal regions during 1998. Methane emissions from the 1998 fires were estimated to be in the range 3.9 to 6.3 Tg CH<sub>4</sub> depending on the characteristics of the fire and types of vegetation burned, with a best estimate of 5.7 Tg CH<sub>4</sub> due to the relatively large amount of peat that burned. Additional CH<sub>4</sub> was emitted by the large fires in Indonesia during late-1997. Levine [1999] used estimates of the area burned, separated it into sub-areas by vegetation type, and estimated a possible range of CH<sub>4</sub> emission of 1.2 to 3.7 Tg CH<sub>4</sub> with a most likely value of 1.8 Tg CH<sub>4</sub>. So, biomass burning, particularly in boreal regions, likely made a small contribution to the anomalous CH<sub>4</sub> increase during 1998.

Reaction of CH<sub>4</sub> with hydroxyl radical (OH) in the troposphere represents about 90% of the total CH<sub>4</sub> sink, and it is the largest term in the global methane budget. The temperature anomalies observed during 1998 have the potential to affect the magnitude of this term. The rate coefficient for the reaction increases by  $\sim 2\%$  for each 1°C increase. Also, assuming constant relative humidity, atmospheric [H<sub>2</sub>O] could have increased with higher tropospheric temperatures, thereby increasing [OH]. In addition, during 1998, the QBO 30 hPa zonal wind index was in an easterly phase, and a negative global anomaly in stratospheric O<sub>3</sub> was observed [Hamilton and Fan, 2000]. Lower stratospheric O<sub>3</sub> by itself would lead to increased tropospheric OH, and lower CH<sub>4</sub> growth rates, opposite to what was observed. We have no evidence from the CH<sub>4</sub> measurements that any of these possible effects on [OH] occurred. The QBO also affects mixing rates between the stratosphere and the troposphere; this could have led to a small increase in CH<sub>4</sub> growth rate of 1-2 ppb yr<sup>-1</sup> [Hamilton and Fan, 2000].

The CH<sub>4</sub> increase in the southern hemisphere in 1999 (+6.90.4 ppb) was considerably larger than in the northern hemisphere, where CH<sub>4</sub> values decreased (-1.6 1.1ppb). A similar pattern was observed in 1988-89; both periods of relatively large CH<sub>4</sub> growth in the southern hemisphere were during strong La Niña events. Since only  $\sim 30\%$  of global emissions occur in the southern hemisphere, it is unlikely that an increase in emissions alone caused such a large change in growth rate. Though we can not rule out that there was a significant decrease in the CH<sub>4</sub> sink in the southern hemisphere during 1998-99, we suggest that a possible contributor to the increase in the southern hemisphere was enhanced interhemispheric exchange as previously described by Steele et al. [1992]. It has been suggested that the meteorological conditions that exist during La Niña events are conducive to enhanced interhemispheric exchange [see e.g., Prinn et al., 1992]. Since the average interhemispheric difference (i.e., the difference between annual mean CH<sub>4</sub> mole fractions, NH-SH) for 1984 to 1999 is 88.8 2.9ppb (where uncertainty is 1\_), enhanced interhemispheric exchange leads to increased CH<sub>4</sub> growth

rate in the southern hemisphere. This suggestion is supported by a decrease in the interhemispheric difference during 1999 to 82.9 ppb, the smallest interhemispheric difference in our CH<sub>4</sub> time series (16 years).

#### 4. Summary and Conclusions

The globally averaged methane growth rate increased from an average of 3.9 ppb yr<sup>-1</sup> during 1995-1997 to 12.7 ppb yr<sup>-1</sup> in 1998. This change in growth rate implies an anomalous imbalance between sources and sinks equal to 24 Tg CH<sub>4</sub> during 1998. During 1999, the global increase was 2.6 ppb, but CH<sub>4</sub> decreased by 1.6 ppb in the northern hemisphere. This suggests that 1998 was an anomaly and not a return to the large growth rates observed during the late-1970s and early-1980s. The spatial distribution of the anomalous increase is an indicator of the sources that contributed. Based on latitude zones comprising 25% of the atmosphere, the largest increases during 1998 were at 30-90°N and 0-30°S. Model calculations suggest there were increased CH<sub>4</sub> emissions from wetlands, contributing an additional 11.5 Tg CH<sub>4</sub> in the NH and 13 Tg CH<sub>4</sub> in the tropics (30°S-30°N), due to a warmer and wetter environment in wetland regions. Large boreal fires, particularly in Siberia, may have also contributed up to 4 Tg CH<sub>4</sub>. Though our model estimates of wetland emission anomalies are likely overestimated, a clear link between CH<sub>4</sub> emissions and climate exists. If wetland regions trend towards warmer wetter environments than they are currently, CH<sub>4</sub> emissions are likely to increase.

#### Figure Captions

Figure 1. Locations of sites in the NOAA CMDL cooperative air sampling network used in this study (circles). Some sites were not active for the entire period (1984-1999). Large circles are "fixed" sites, and small circles are approximate samplings locations along ship's cruise tracks. The gray-shaded areas identify wetland regions [based on *Matthews and Fung, 1987*].

Figure 2. (a) Globally averaged CH<sub>4</sub> mole fractions. Solid line is the deseasonalized trend (T(t); see text). (b) Globally averaged instantaneous growth rate determined as the time-derivative of the trend line in (a). Symbols are global annual increases in Table 1. Uncertainties are 1\_ determined with a nonparametric statistical technique [*Dlugokencky et al., 1994b*].

**Table 1. Annual increases in atmospheric CH<sub>4</sub> by latitude zone (ppb).**

YEAR	Latitude Zone						Global	Predict <sup>1</sup>
	30-90°S	0-30°S	0-30°N	30-90°N	0-90°S	0-90°N		
1984	11.9 0.8	14.4 0.5	11.7 2.3	17.2 2.2	13.1 0.5	14.4 1.6	13.8 0.9	14.5
1985	11.9 0.6	11.2 0.4	13.9 2.3	10.3 2.6	11.6 0.4	12.1 1.7	11.8 0.9	13.3
1986	12.0 0.7	10.2 1.0	13.9 1.9	14.3 1.5	11.1 0.7	14.1 1.4	12.6 0.9	12.2
1987	9.8 0.4	9.1 0.7	11.0 1.6	14.4 1.9	9.5 0.4	12.7 1.4	11.1 0.8	11.2
1988	12.7 0.3	15.4 0.8	7.4 1.2	6.7 1.2	14.0 0.4	7.0 0.8	10.5 0.5	10.3
1989	11.4 0.7	8.6 0.7	11.9 1.1	10.2 1.3	10.0 0.6	11.1 1.0	10.5 0.6	9.46
1990	6.3 0.9	7.0 0.6	10.1 1.1	10.9 1.0	6.6 0.5	10.5 0.7	8.6 0.5	8.7
1991	15.8 0.7	15.1 0.4	15.4 1.4	16.0 1.5	15.4 0.4	12.7 1.1	14.1 0.6	7.9
1992	6.4 0.3	4.0 0.4	-0.7 1.0	0.1 1.1	5.2 0.2	-0.3 0.8	2.5 0.4	7.3
1993	0.3 0.7	3.2 0.4	6.1 1.0	7.3 0.8	1.7 0.4	6.7 0.7	4.2 0.4	6.7
1994	7.7 0.5	8.2 0.3	7.7 1.6	5.1 0.9	8.0 0.3	6.5 1.0	7.2 0.5	6.1
1995	4.4 1.0	4.6 0.4	1.5 1.5	2.9 0.8	4.5 0.6	2.2 0.9	3.3 0.5	5.6
1996	3.0 0.8	2.4 0.5	4.3 0.9	1.9 0.7	2.7 0.5	3.1 0.6	2.9 0.4	5.2
1997	6.4 0.4	3.3 0.6	6.7 2.0	5.1 1.2	4.9 0.3	5.9 1.1	5.4 0.6	4.7
1998	11.1 0.2	14.0 0.8	10.5 1.8	15.2 1.2	12.6 0.4	12.8 1.1	12.7 0.6	4.3
1999	7.2 0.7	6.6 0.4	0.0 1.0	-3.2 2.0	6.9 0.4	-1.6 1.1	2.6 0.6	4.0

Uncertainties are 1<sub>σ</sub> determined with a nonparametric statistical technique [Steele *et al.*, 1992].

<sup>1</sup>Predicted global increase based on constant CH<sub>4</sub> emissions and lifetime [Dlugokencky *et al.*, 1998].

**Table 2. Methane emission anomalies from natural wetlands by semihemisphere calculated with the regression model.**

YEAR	Model Emission Anomaly (Tg CH <sub>4</sub> )		
	30-90°N	Equ-30°N	Equ-30°S
1998	+11.6	+1.5	+11.5
1999	-7.9	-5.7	-4.2

## References

- Bekki, S., K.A. Law, and J.A. Pyle, Effect of ozone depletion on atmospheric CH<sub>4</sub> and CO, *Nature*, 371, 595-597, 1994.
- Dlugokencky, E.J., K.A. Masarie, P.M. Lang, P.P. Tans, L.P. Steele, and E.G. Nisbet, A dramatic decrease in the growth rate of atmospheric methane in the northern hemisphere during 1992, *Geophys. Res. Lett.*, 21, 45-48, 1994a.
- Dlugokencky, E.J., L.P. Steele, P.M. Lang, and K.A. Masarie, The growth rate and distribution of atmospheric methane, *J. Geophys. Res.*, 17,021-17,043, 1994b.
- Dlugokencky, E.J., E.G. Dutton, P.C. Novelli, P.P. Tans, K.A. Masarie, K.O. Lantz, and S. Madronich, Changes in CH<sub>4</sub> and CO growth rates after the eruption of Mt. Pinatubo and their link with changes in tropical tropospheric UV flux, *Geophys. Res. Lett.*, 23, 2761-2764, 1996.
- Dlugokencky, E.J., K.A. Masarie, P.M. Lang, and P.P. Tans, Continuing decline in the growth rate of the atmospheric methane burden, *Nature*, 393, 447-450, 1998.
- Dutton, E.G. and J.R. Christy, Solar radiative forcing at selected locations and evidence for global lower tropospheric cooling following the eruptions of El Chichón and Pinatubo, *Geophys. Res. Lett.*, 19, 2313-2316, 1992.
- Etheridge, D.M., L.P. Steele, R.J. Francey, and R.L. Langenfelds, Atmospheric methane between 1000 A.D. and present: Evidence of anthropogenic emissions and climate variability, *J. Geophys. Res.*, 103, 15,979-15,993, 1998.
- Francey, R. J., M.R. Manning, C.E. Allison, S.A. Coram, D.M. Etheridge, R.L. Langenfelds, D.C. Lowe, and L.P. Steele, A history of <sup>13</sup>C in atmospheric CH<sub>4</sub> from the Cape Grim Air Archive and Antarctic firm air, *J. Geophys. Res.*, 104, 23,631-23,643, 1999.
- Fung, I., J. John, J. Lerner, E. Matthews, M. Prather, L.P. Steele, and P.J. Fraser, Three-dimensional model synthesis of the global methane cycle, *J. Geophys. Res.*, 96, 13,033-13,065, 1991.
- Hamilton, K. and S. Fan, Effects of the stratospheric QBO on long-lived greenhouse gases in the troposphere, *J. Geophys. Res.*, in press, 2000.
- Hansen, J., R. Ruedy, J. Glascoe, and M. Sato, GISS analysis of surface temperature change, *J. Geophys. Res.*, 104, 30,997-31,022, 1999.
- Hogan, K.B. and R.C. Harriss, Comment on 'A dramatic decrease in the growth rate of atmospheric methane in the northern hemisphere during 1992' by E.J. Dlugokencky et al., *Geophys. Res. Lett.*, 21, 2445-2446, 1994.
- Karlsdóttir, S and I.S.A. Isaksen, Changing methane lifetime: Possible cause for reduced growth, *Geophys. Res. Lett.*, 27, 93-96, 2000.
- Kasischke, E.S, K. Bergen, R. Fennimore, F. Sotelo, G. Stephens, A. Janetos, and H.H. Shugart, Satellite imagery gives a clear picture of Russia's boreal forest fires, *EOS Transactions of the American Geophysical Union*, 80, 141 and 147, 1999
- Khalil, M.A.K., R.A. Rasmussen, M.J. Shearer, R.W. Dalluge, L. Ren, and C.-L. Duan, Factors affecting methane emissions from rice fields, *J. Geophys. Res.*, 103, 25,219-25,231, 1998.
- Krol, M, P.J. van Leeuwen, and J. Lelieveld, Global OH trend inferred from methylchloroform measurements, *J. Geophys. Res.*, 103, 10,697-10,711, 1998.
- Law, K.S. and E.G. Nisbet, Sensitivity of the CH<sub>4</sub> growth rate to changes in CH<sub>4</sub> emissions from natural gas and coal, *J. Geophys. Res.*, 101, 14,387-14,397, 1996.
- Levine, J., The 1997 fires in Kalimantan and Sumatra, Indonesia: Gaseous and particulate emission, *Geophys. Res. Lett.*, 26, 815-818, 1999.
- Matthews, E. and I. Fung, Methane emission from natural wetlands: Global distribution, area, and environmental characteristics of sources, *Global Biogeochem. Cycles*, 1, 61-86, 1987.
- Prinn, R., D. Cunnold, P. Simmonds, F. Alyea, B. Boldi, A. Crawford, P. Fraser, D. Gutzler, D.

- Hartley, R. Rosen, and R. Rasmussen, Global average concentration and trend for hydroxyl radicals deduced from ALE-GAGE trichloroethane (methyl chloroform) data for 1978-1990, *J. Geophys. Res.*, 97, 2445-2461, 1992.
- Steele, L.P., E.J. Dlugokencky, P.M. Lang, P.P. Tans, R.C. Martin, and K.A. Masarie, Slowing down of the global accumulation of atmospheric methane during the 1980s, *Nature*, 358, 313-316, 1992.
- Thoning, K.W., P.P. Tans, and W.D. Komhyr, Atmospheric carbon dioxide at Mauna Loa Observatory, 2. Analysis of the NOAA GMCC data, 1974-1985, *J. Geophys. Res.*, 94, 8549-8565, 1989.
- Walter, B.P., Development of a process-based model to derive methane emissions from natural wetlands for climate studies, Dissertation, Examensarbeit 60, Max-Planck-Institut für Meteorol., Hamburg, Germany, 1998.
- Walter, B.P., and M. Heimann, A process-based, climate sensitive model to derive methane emissions from natural wetlands: Applications to five wetland sites, sensitivity to model parameters and climate, *Global Biogeochem. Cycles*, in press, 2000.

